Application of Lindhard's dielectric theory to the stopping of ions in solids

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Lindhard's dielectric theory of energy loss is applied to the stopping of energetic ions in solids. The Lindhard formalism, in combination with the local density approximation and the concept of effective charge, is used to calculate the stopping powers of heavy ions in many targets and over a wide range of projectile energies. Theoretically calculated stopping powers are compared with available experimental data.

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INTRODUCTION

For many years, the stopping of energetic ions in matter has been a subject of great interest to theoretical and experimental physicists. Bohr,[1-3] Bethe,[4-6] and Bloch[7,8] were the pioneering theoretical investigators of charged particle stopping in matter. Some 30 years after the development of Bethe-Bloch theory, Lindhard[9] presented an elegant dielectric formulation of stopping theory for a charged particle penetrating a free-electron gas.

The Lindhard dielectric formalism probably stands as the most complete "first principles" theory of energy loss to date. However, the application of Lindhard's theory to real solids becomes a formidable task since the direct numerical integration of the Lindhard stopping equation is slow and cumbersome due to the presence of singularities in the integrand. This paper represents the first attempt to apply, with precision, the Lindhard theory of stopping to actual solids.

The original work of Lindhard was extended by Lindhard and Winther[10] (L-W) who obtained an analytic expansion for the stopping power in the limit of high and low projectile velocity relative to the Fermi velocity of an electron gas; the Bethe stopping power (with a redefined mean ionization potential) plus shell corrections were obtained in the high-velocity limit whereas a linear dependence of velocity with stopping power was shown at low velocity.

Bonderup[11] utilized the L-W expansions to include the local density approximation of Lindhard and Scharff[12] (L-S) so as to extract the Bethe-Bloch terms, commonly referred to as the mean ionization potential and the shell corrections. In the local density approximation, each volume element of the solid is considered to be an independent plasma of uniform density, so that the total electronic stopping power is obtained by spatially averaging a specific stopping power over the target electron charge distributions. Bonderup, in his calculations, uses the Lenz-Jensen atomic charge distribution to describe the target electrons. His calculation was subsequently refined by Rousseau, Chu, and Powers[13] (RCP) who utilized Hartree-Fock-Slater[14] (HFS) charge distributions to describe the spatially varying electron density, thereby predicting the presence of oscillations in the stopping power when plotted versus target atomic number.

In this paper, we evaluate the Lindhard dielectric theory of stopping power within the local density approximation and the concept of effective charge. A comparison of Lindhard's stopping powers with the experiment for 0.4-MeV/amu heavy ions in solid targets is given; comparative agreement between theory and experiment over a wide range of projectile energies is discussed.

LINDHARD'S THEORY OF STOPPING POWER[9]

For an ion of charge ze moving with velocity v in a medium of uniform density ρ, the energy loss due to electron excitation can conveniently be written in the form

$$\left(-\frac{dE}{dx}\right)=\frac{4\pi}{m}\left(\frac{ze^2}{v}\right)^2\rho L(\rho,v), \tag{1}$$

where L is the stopping number and m is the mass of the electron. In the dielectric formalism, L is written as

$$L=\frac{i}{\pi\omega_0^2}\int_0^{\infty}\frac{dk}{k}\int_{kv}^{\infty}\omega\,d\omega[\epsilon^{-1}(k,\omega)-1], \tag{2}$$

where ω_0 is the plasma frequency, i.e.,

$$\omega_0^2=\frac{4\pi e^2\rho}{m}, \tag{3}$$

and $\epsilon(k,\omega)$ is the wave number and frequency dependent longitudinal dielectric constant.

Lindhard obtained the dielectric constant for a free-electron gas within first-order perturbation theory as

$$\epsilon(k,\omega)=1+\frac{8\pi me^2}{h^2k^2}\sum_n F(E_n)\times\left(\frac{1}{k^2+2\mathbf{k}\cdot\mathbf{k}_n-(2m/h^2)(\omega+i\gamma)}+\frac{1}{k^2-2\mathbf{k}\cdot\mathbf{k}_n+(2m/h^2)(\omega+i\gamma)}\right), \tag{4}$$

where E_n and $\mathbf{k}_n$ represent the energy and wave vector for an electron in the nth state and γ is a small positive value. For a free-electron gas at zero temperature,

$$F(E_n)=1 \quad \text{for } E_n<E_F,$$
$$F(E_n)=0 \quad \text{for } E_n>E_F, \tag{5}$$

and the summation in Eq. (4) can be evaluated to get

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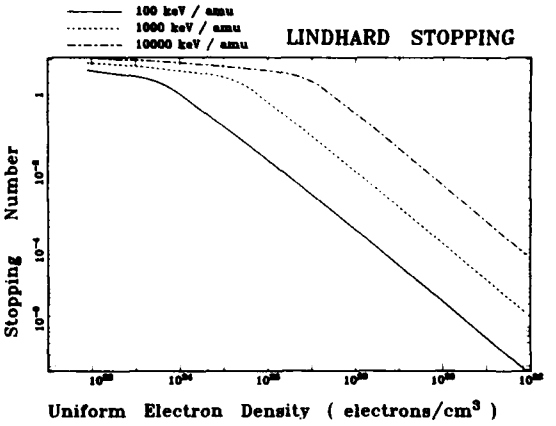

FIG. 1. Plot of variation in stopping number [Eq. (11)] with electron density for select projectile energies.

$$\epsilon(u,z) = 1 + (\chi^2/z^2)[f_1(u,z) + if_2(u,z)], \tag{6}$$

where

$$f_1(u,z) = \tfrac{1}{2} + (1/8z)[1-(z-u)^2] \times \ln|(z-u+1)/(z-u-1)| + (1/8z)[1-(z+u)^2] \times \ln|(z+u+1)/(z+u-1)|, \tag{7a}$$

and

$$f_2(u,z) = \tfrac{1}{2}\pi u, \quad \text{for } z+u<1$$

$$f_2(u,z) = \frac{\pi}{8z}[1-(z-u)^2], \quad \text{for } |z-u|<1<z+u$$

$$f_2(u,z) = 0, \quad \text{for } |z-u|>1. \tag{7b}$$

Here the variables k and ω have been replaced by reduced variables

$$z = (k/2k_F) \text{ and } u = (\omega/kv_F), \tag{8}$$

where k_F and v_F are defined by

$$E_F = \tfrac{1}{2}mv_F^2 \equiv (h^2k_F^2/2m) = (h^2/2m)(3\pi^2\rho)^{2/3}. \tag{9}$$

In addition, the parameter χ^2 in Eq. (6) is defined by

$$\chi^2 = (v_0/\pi v_F), \tag{10}$$

where $v_0 = (e^2/h)$, the Bohr velocity. When the dielectric constant of Eq. (6) is substituted into Eq. (2), one obtains

$$L = \frac{6}{\pi}\int_0^{v/v_F} u\,du \times \int_0^{\infty} dz \frac{z^3 f_2(u,z)}{[z^2+\chi^2 f_1(u,z)]^2 + [\chi^2 f_2(u,z)]^2}. \tag{11}$$

This expression for L in Eq. (11) is the Lindhard stopping number. In this paper, we make use of stopping numbers obtained from a numerical integration of Eq. (11) as a continuous function of electron density (or χ^2) over a wide range of projectile velocities (or energies). While the detailed results of the calculation can be found elsewhere,[15] some typical illustrations of the variation in stopping number with electron density and energy can be found in Fig. 1.

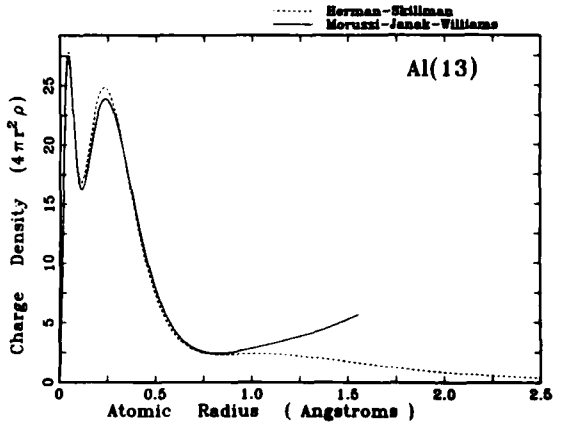

FIG. 2. Comparison of HFS radial charge density calculated by F. Herman and S. Skillman (Ref. 14) and solid-state spherically averaged radial charge densities by Moruzzi, Janak, and Williams (Ref. 16).

LOCAL DENSITY APPROXIMATION

In the local density approximation,[12] each volume element of the solid is considered to be an independent plasma of uniform density ρ, so that the total electronic stopping power is given by

$$\left(-\frac{dE}{dx}\right) = \frac{4\pi}{m}\left(\frac{ze^2}{v}\right)^2 \int_0^{\infty} \rho(r)\, L(\rho,v)\, 4\pi r^2\, dr, \tag{12}$$

where $\rho(r)$ is the spherically averaged charge density of the target atom. In this paper we use spherically averaged solid-state charge densities[16] in evaluating Eq. (12). Figure 2 shows a comparison of the HFS radial charge density calculated by F. Herman and S. Skillman[14] and solid-state spherically averaged radial charge densities calculated by Moruzzi, Janak, and Williams[16]; there is a pronounced difference in the spatial variation of the solid-state and atomic charge densities away from the interior of the target atom due to solid-state bonding effects. This spatial variation in charge density

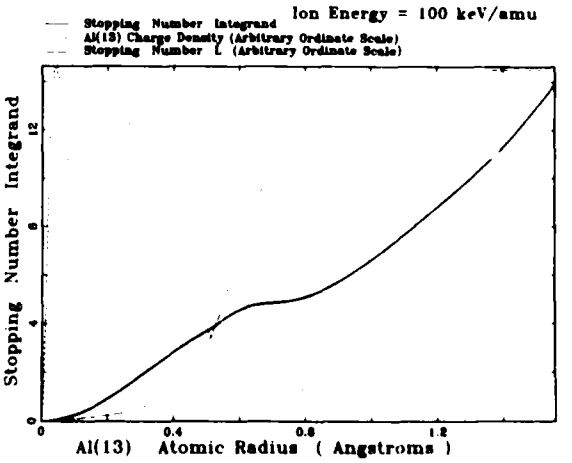

FIG. 3. Comparison of spatial variation in the stopping number integrand of Eq. (12), the stopping number of Eq. (12), and the solid-state radial charge density for aluminum (Ref. 16) with ion energy of 100 keV/amu.

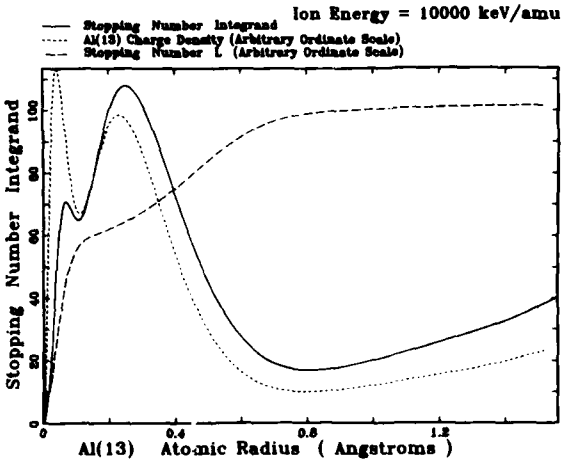

FIG. 4. Comparison of spatial variation in the stopping number integrand of Eq. (12), the stopping number of Eq. (12), and the solid-state radial charge density for aluminum (Ref. 16) with ion energy of 10 000 keV/amu.

gives rise to a marked change in density-averaged stopping number. Inspection of Figs. 3 and 4 shows that the stopping number integrand of Eq. (12) approximately follows the radial charge density in spatial variation. It should also be noted that the low-energy (100 keV/amu) projectile stopping number is influenced mainly by the outer shell electron charge distribution, whereas the high-energy projectile (10 000 keV/amu) stopping number is dependent upon the inner electron shells as well. This analysis clearly shows the importance of using solid-state charge densities in the calculation of stopping powers of solids.

EFFECTIVE CHARGE OF ENERGETIC IONS IN SOLIDS

In the preceding discussions, the projectile ion was assumed to be a bare nuclear point charge. However, when the projectile is an ionized atom with an intrinsic electronic charge distribution, the question of the net charge of the projectile arises. From the beginning of stopping power theory the charge of heavy ions in a solid has been poorly understood. Bohr[1] suggested that the ion might be assumed to be stripped of all electrons with orbital velocities less than the ion velocity. This concept is difficult to calculate in detail for heavy ions because of the difficulties in assigning velocities to the highly distorted electron orbitals of a partially stripped ion in a dense electron plasma.

The active interest in fission fragments in 1939–1941 brought about various attempts to predict heavy ion stopping powers using the concept of effective charge.[17-20] Within this concept, the ion charge state was assumed to be independent of the ion-solid stopping interaction so that the ion-target stopping interaction chould be represented by an experimental proton stopping power, and the ratio of a heavy ion stopping power to the proton value (for the same velocity in the material) was identified as the heavy ion effective charge (it was assumed that the proton charge was unity). This quantity included not only the charge state of the ion, but also all reduction in stopping due to screening effects and other perturbations. (See review by Brandt for theoretical details.[21]) Remarkably, this quantity, for all heavy ions in all targets over several decades of energy, can be represented by a simple empirical formula established by Northcliffe[22] as

$$z^*/z = 1 - \exp(v/v_0 z^{2/3}), \tag{13}$$

where z^* is the effective charge and $v_0 = e^2h$, is the Bohr velocity ($2.188 \cdot 10^8$ cm/sec). This formula has been reshaped by many authors as thousands of heavy ion stopping powers have been measured. We use the form[23]

$$z^*/z = 1 - [\exp(-A)] \times [1.034 - 0.1777\ \exp(-0.08114z)], \tag{14}$$

where

$$A = B + 0.0378 \sin\tfrac{1}{2}\pi B, \qquad B = 0.886(40E/m)^{1/2}/z^{2/3}, \tag{15}$$

where the ion energy E is in MeV, and the ion mass is in amu. The expression is considered to be accurate *to about* 5% for ions with atomic numbers 6–92 in solids with atomic numbers 4–79, for energies above 0.2 MeV/amu. This formula is independent of target and depends only on the ion atomic number and velocity.

COMPARISON OF THEORY WITH EXPERIMENT: CONCLUSIONS

Within the local density approximation and the concept of effective charge, we have applied the Lindhard stopping theory for a charged particle in a free-electron gas to the stopping of heavy ions in solids. Stopping powers [Eq. (12) with z replaced by z^*] were calculated for a wide variety of projectile energies ranging from 100 to 10 000 keV/amu and for many targets. For projectile energies of 4000 keV/amu and above, the *maximum error* incurred in comparing the calculated stopping powers with experiment[24] for many targets is about 4%. For projectile energies below 4000 keV/amu, the maximum error increases monotonically to a value of about 30% for a projectile energy of 100 keV/amu. A typical comparison of theoretical and experimental stopping powers for many targets is illustrated in Fig. 5. Here the ratio of experimental to theoretical stopping is plotted versus

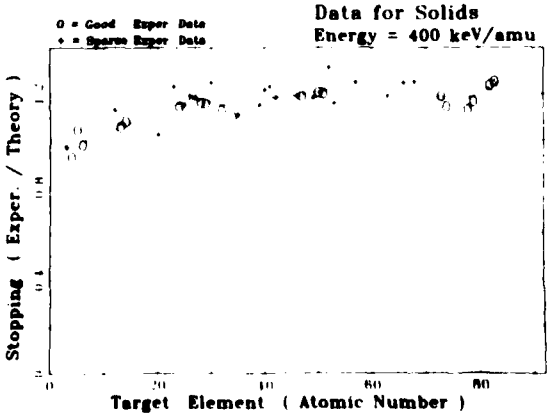

FIG. 5. Plot of the ratio of experimental (Ref. 24) to theoretical stopping versus target atomic number for projectile energy of 400 keV/amu.

target atomic number for a projectile energy of 400 keV/amu.

The objective of this paper has been to apply the Lindhard theory of stopping to real solids. Within the assumed approximations, the results show that the theory significantly deviate from experiment in the low projectile energy (100 keV/amu$<E<$4000 keV/amu) region. The reasons for this disagreement may be manyfold: the use of a free-electron gas dielectric response function, the extrapolation of the uniform electron gas model to solid state systems via the local density approximation, and the use of the effective charge concept to reduce experimental stopping powers to stopping numbers. In a future paper, we plan to explore an extension of Lindhard's theory to self-consistently include heavy atom projectiles which may abrogate some of the above weaknesses.

[1]N. Bohr, Philos. Mag. **25**, 10 (1913).
[2]N. Bohr, Philos. Mag. **30**, 581 (1915).
[3]N. Bohr, Mat. Fys. Medd. Dan. Vid. Selsk. **18**, 1 (1948).
[4]H.A. Bethe, Ann. Phys. **5**, 325 (1930).
[5]H.A. Bethe, Z. Phys. **76**, 293 (1932).
[6]H.A. Bethe and W. Heitler, Proc. R. Soc. London, **A146**, 83 (1934).
[7]F. Bloch, Ann. Phys. **16**, 287 (1933).
[8]F. Bloch, Z. Phys. **81**, 363 (1933).
[9]J. Lindhard, Mat. Fys. Medd. Dan. Vid. Selsk. **28**, 1 (1954).
[10]J. Lindhard and A. Winther, Mat. Fys. Medd. Dan. Vid. Selsk. **34**, 1 (1964).
[11]E. Bonderup, Mat. Fys. Medd. Dan. Vid. Selsk. **35**, 1 (1967).
[12]J. Lindhard and M. Scharff, Mat. Fys. Medd. Dan. Vid. Selsk. **27**, 1 (1953).
[13]C.C. Rousseau, W.K. Chu, and D. Powers, Phys. Rev. **A4**, 1066 (1970).
[14]F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs, 1963).
[15]G.J. Iafrate and J.F. Ziegler, J. Appl. Phys. **50**, 5579 (1979).
[16]V.L. Moruzzi, J.F. Janak, and A.R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).
[17]W.E. Lamb, Phys. Rev. **58**, 696 (1940).
[18]N. Bohr, Phys. Rev. **59**, 270 (1941).
[19]J. Knipp and E. Teller, Phys. Rev. **59**, 659 (1941).
[20]J.H.M. Brunings, J. Knipp, and E. Teller, Phys. Rev. **60**, 657 (1941).
[21]W. Brandt, in *Atomic Collisions in Solids* (Plenum, New York, 1975), p. 261.
[22]L.C. Northcliffe, Phys. Rev. **120**, 1744 (1961).
[23]J.F. Ziegler, Appl. Phys. Lett. **31**, 544 (1977).
[24]J.F. Ziegler, *The Stopping and Range of Ions in Matter* (Pergamon, New York, 1979), Vol. 5.